Materials Chemomechanics by Inference-Boosted First Principles Modelling

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Many technologically important phenomena coupling complex chemistry with high-gradient elastic fields such as thermally activated fracture [1], or stress corrosion [2] of brittle materials are beyond the reach of First-Principles Molecular Dynamics (FPMD)-techniques, whether based on standard or O(N) implementations. This is ultimately because the required model system sizes are too large, and/or the required simulation times are too long, for these high-accuracy traditional approaches. In most (but not all) situations, using classical MD is not a viable alternative, as suitably general and accurate reactive force fields are not available, nor are fitting databases a priori guaranteed to contain the information necessary to describe all the chemical processes encountered along the dynamics. The problem, widespread in covalent/brittle systems, can arise in ductile metallic systems too [3]. Finally, QM/MM techniques combining quantum and classical zones in a single calculation also pose significant difficulties, especially when the target processes involving sustained mass transport into and out of the (e.g., fast moving) QM zone.

This situation strongly calls for the use of big-data based MD techniques, capable of locating and using QM-accurate information pre-stored in massive databases, or generating it if no such information is available. In practical realisations, these could be configuration databases dynamical coupled with specially-tuned Machine Learning force fields which minimise the computational workload of MD runs by allowing QM subroutine calls only when chemically novel configurations are encountered along the systems trajectory. I will present one such Learn On the Fly technique, effectively unifying First-Principles Molecular Dynamics and Machine Learning into a single, information efficient simulation scheme capable of learning/predicting atomic forces through Bayesian inference [4]. Recently developed covariant kernels specifically designed for direct force learning by Gaussian Process regression are at the core of this technique [5]. Interestingly, QM-zone partitioning approaches followed by execution via any of the existing O(N^3) QM engines is predicted to be a better option than using O(N) QM methods when dealing with large QM zones in QM/MM calculations running on high-end parallel platforms [6-7].

[1] J.R.Kermode et al., PRL 115, 135501 (2015).

[2] A. Gleizer et al., PRL 112, 115501 (2014).

[3] F.Bianchini et al., Mod. and Simul. in Mat. Science and Engineering 24, 045012 (2016).

[4] Z. Li, J. R. Kermode and A. De Vita, Phys. Rev. Lett., 114, 096405 (2015).

[5] A.Glielmo and A. De Vita, in preparation.

[6] Cf., e.g., the US-DOE INCITE on SiO2 ML-Fracture Project https://www.alcf.anl.gov/projects/sio2-fracture-chemomechanics-machine-learning-hybrid-qmmm-scheme

[7] M. Caccin et al., Int. J. of Quantum Chemistry 2015, DOI: 10.1002/qua.24952